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# United States Patent [19]

Berlowitz

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[54] **ADDITIVE COMBINATION TO REDUCE DEPOSIT FORMING TENDENCIES AND IMPROVE ANTIOXIDANCY OF AVIATION TURBINE OILS (LAW406)**

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[51] Int. Cl.<sup>6</sup> ..... **C09K 15/26**; C09K 3/00; C23F 11/00

[52] U.S. Cl. .... **252/399**; 252/391; 252/394; 252/395; 252/402; 508/258; 508/570

[58] Field of Search ..... 252/399, 402, 252/391, 394, 395; 508/258, 570

3,573,206	3/1971	Braid et al. ....	252/51.5
3,609,080	9/1971	Dazzi .....	252/47.5
3,642,630	2/1972	MacPhail et al. ....	252/47.5
3,759,996	9/1973	Braid .....	260/576
3,773,665	11/1973	Braid .....	252/50
4,081,386	3/1978	Sowerby .....	508/258
4,280,894	7/1981	Taylor .....	208/15
4,292,232	9/1981	Dazzi et al. ....	260/408
4,737,300	4/1988	Wirth et al. ....	252/41
4,820,756	4/1989	Pitteloud et al. ....	524/289
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4,931,196	6/1990	Payne et al. ....	252/47.5
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5,176,850	1/1993	O'Neill .....	252/395
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1287647	9/1972	United Kingdom .....	C10M 3/40

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2,657,982	11/1953	Hill et al. ....	44/63
3,198,797	8/1965	Dexter et al. ....	260/249.5
3,250,708	5/1966	Dazzi et al. ....	252/28
3,255,191	6/1966	Dexler et al. ....	260/248
3,278,436	10/1966	Dazzi et al. ....	252/50
3,322,763	5/1967	Dazzi et al. ....	260/249.6
3,492,233	1/1970	Heppiewhite et al. ....	252/51.5
3,509,214	4/1970	Braid et al. ....	260/576

### [57] ABSTRACT

An aviation turbine oil of reduced deposit forming tendencies and improved anti-oxidancy is disclosed which comprises a major portion of a suitable aviation turbine oil base stock and a minor amount of a non-sulfur containing triazine derivative and a mercapto alkyl alcohol.

**10 Claims, No Drawings**

**ADDITIVE COMBINATION TO REDUCE  
DEPOSIT FORMING TENDENCIES AND  
IMPROVE ANTIOXIDANCY OF AVIATION  
TURBINE OILS (LAW406)**

**BACKGROUND OF THE INVENTION**

**1. Field of the Invention**

This invention relates to ester-based, in particular diester and polyol ester-based turbo oils which exhibit superior antioxidant and reduced deposit forming tendencies. More particularly it is related to turbo oils comprising esters of pentaerythritol with fatty acids as basestock, and containing a combination of additives which impart improved antioxidant and reduced deposit formation.

**2. Description of the Related Art**

Organic compositions such as mineral oils and lubricating compositions are subject to deterioration by oxidation and in particular are subject to such deterioration at high temperatures in the presence of air. This deterioration often leads to buildup of insoluble deposits which can foul engine parts, deteriorate performance, and increase maintenance. This is particularly the case for lubricating oils used in jet aircraft where wide temperature ranges and extreme operating conditions are likely to be encountered. Proper lubrication of aircraft gas turbines, for example, requires the ability to function at bulk oil temperatures as low as  $-65^{\circ}$  F. to as high as  $450^{\circ}$ – $500^{\circ}$  F.

Most lubricants contain additives to inhibit their oxidation. For example, U.S. Pat. No. 3,773,665 discloses a lubricant composition containing an antioxidant additive mixture of dioctyl diphenylamine and a substituted naphthylamine. U.S. Pat. Nos. 3,759,996; 3,573,206; 3,492,233, and 3,509,214 disclose various methods of oxidatively coupling alkylated diphenylamines with substituted naphthylamines.

Patents disclosing the use of tri-substituted triazines in lubricants generally demonstrate the antioxidant function of these molecules when either used alone, or in combination with other antioxidants. They do not describe the use of these materials as anti-deposition additives. U.S. Pat. No. 3,250,708 describes the use of several triazine derivatives, and combinations with hydroxyl aromatic co-antioxidants. U.S. Pat. Nos. 3,278,436 and 3,322,763 describes tri-substituted triazines including piperidinyll bridged triazines in combination with hydroxyl aromatics.

European Patent application 002,269 discloses the use of tri-substituted triazines where at least one of the amino substituents contains at least one hydrogen as antioxidants, and in combination with arylamine antioxidants.

U.S. Pat. No. 3,642,630 discloses the use of symmetrical and asymmetrical substituted triazines with N-substituted phenothiazine imparts good oxidation stability to synthetic ester based lubricants over a wide range of temperatures.

Other triazine derivatives disclosed in a number of patents to stabilize oils would not be suitable for use in aviation turbine oils as these derivatives contain halogens which are corrosive to metals. For example, U.S. Pat. No. 3,198,797 utilizes 2,4-dichloro-6-dialkyl-dihydroxy-anilino-1,3,5 triazines. Similarly, U.S. Pat. No. 3,202,681 utilizes monohalogen substituted triazines, especially monochloro substituted ones.

It has now been discovered that the deposit forming tendencies and antioxidant properties of these basic antioxidant systems, e.g., tri-substituted triazines alone or in combination with arylamines, can be greatly enhanced by the

addition of a small amount of a sulfur containing additive, specifically mercapto alkyl alcohols (MAA).

**SUMMARY OF THE INVENTION**

The present invention resides in a turbo oil composition exhibiting enhanced antioxidant and resistance to deposit formation, and to a method for achieving that result in turbo oils.

The gas turbine lubricating oil of the present invention comprises a major proportion of synthetic polyol ester based base stock including diesters and polyol esters, preferably polyol ester based base stock and a minor proportion of an antioxidant/deposit control additive comprising a non-sulfur containing, triazine derivative antioxidant and a mercapto alkyl alcohol (MAA). Other, conventional additives such as extreme pressure, pour point reduction, oxidative stability, anti-foaming, hydrolytic stability, improved viscosity index performance, anti-wear, and corrosion inhibitor additives and others may also be employed.

Improved oxidation and deposit control performance in turbo lube oils is achieved by adding to the synthetic polyol ester based lubricating oil an additive package containing a mixture of a non-sulfur containing triazine antioxidant and MAA derivative.

The non-sulfur containing triazine antioxidant is used in an amount in the range 0.1 to 1.2 percent by weight, preferably 0.2 to 0.9 percent, most preferably 0.4 to 0.7 percent, while the MAA is used in an amount in the range 50 to 2000 ppm, preferably 100 to 600 ppm, most preferably 200 to 500 ppm.

The non-sulfur containing triazine antioxidant and the mercapto alkyl alcohol are used in a ratio in the range of 2:1 to 100:1, preferably 4:1 to 40:1, most preferably 5:1 to 20:1.

The use of a non-sulfur containing triazine antioxidant and MAA mixture produces a turbo oil exhibiting markedly superior oxidation and deposit control properties performance as compared to the performance exhibited without the combination.

**DETAILED DESCRIPTION**

A turbo oil having Unexpectedly superior deposition performance comprises a major portion of a synthetic polyol ester base oil and minor portion of an anti-deposition additive package comprising of a mixture of a non-sulfur containing substituted triazine derivative with an MAA. Synthetic esters include diesters and polyol esters.

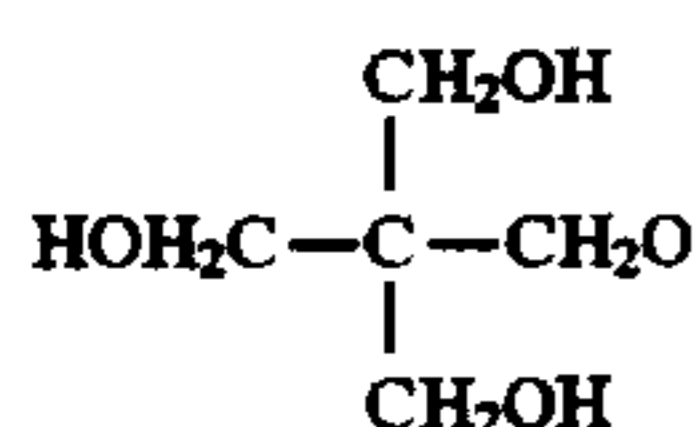
The diesters that can be used for the improved deposition turbo oil of the present invention are formed by esterification of linear or branched  $C_6$ – $C_{15}$  aliphatic alcohols with one of such dibasic acids as adipic, sebacic, or azelaic acids. Examples of diesters are di-2-ethylhexyl sebacate and dioctyl adipate.

The synthetic polyol ester base oil is formed by the esterification of an aliphatic polyol with carboxylic acid. The aliphatic polyol contains from 4 to 15 carbon atoms and has from 2 to 8 esterifiable hydroxyl groups. Examples of polyol are trimethylolpropane, pentaerythritol, dipentaerythritol, neopentyl glycol, tripentaerythritol and mixtures thereof.

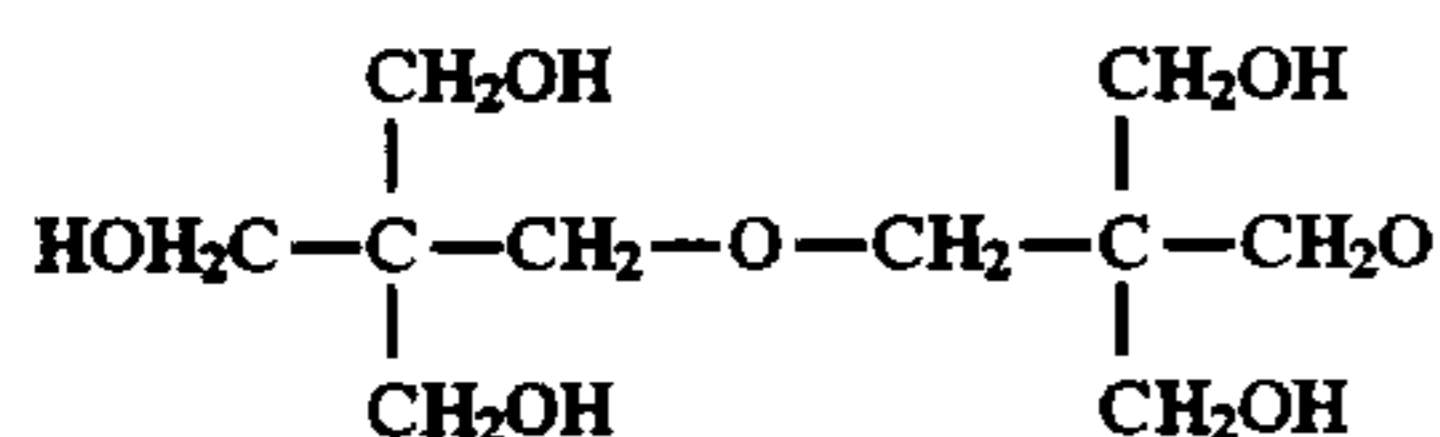
The carboxylic acid reactant used to produce the synthetic polyol ester base oil is selected from aliphatic monocarboxylic acid or a mixture of aliphatic monocarboxylic acid and aliphatic dicarboxylic acid. The carboxylic acid contains from 4 to 12 carbon atoms and includes the straight and branched chain aliphatic acids, and mixtures of monocarboxylic acids may be used.

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The preferred polyol ester base oil is one prepared from technical pentaerythritol and a mixture of C<sub>4</sub>-C<sub>12</sub> carboxylic acids. Technical pentaerythritol is a mixture which includes about 85 to 92% monopentaerythritol and 8 to 15% dipentaerythritol. A typical commercial technical pentaerythritol contains about 88% monopentaerythritol having the formula



and about 12% of dipentaerythritol having the formula



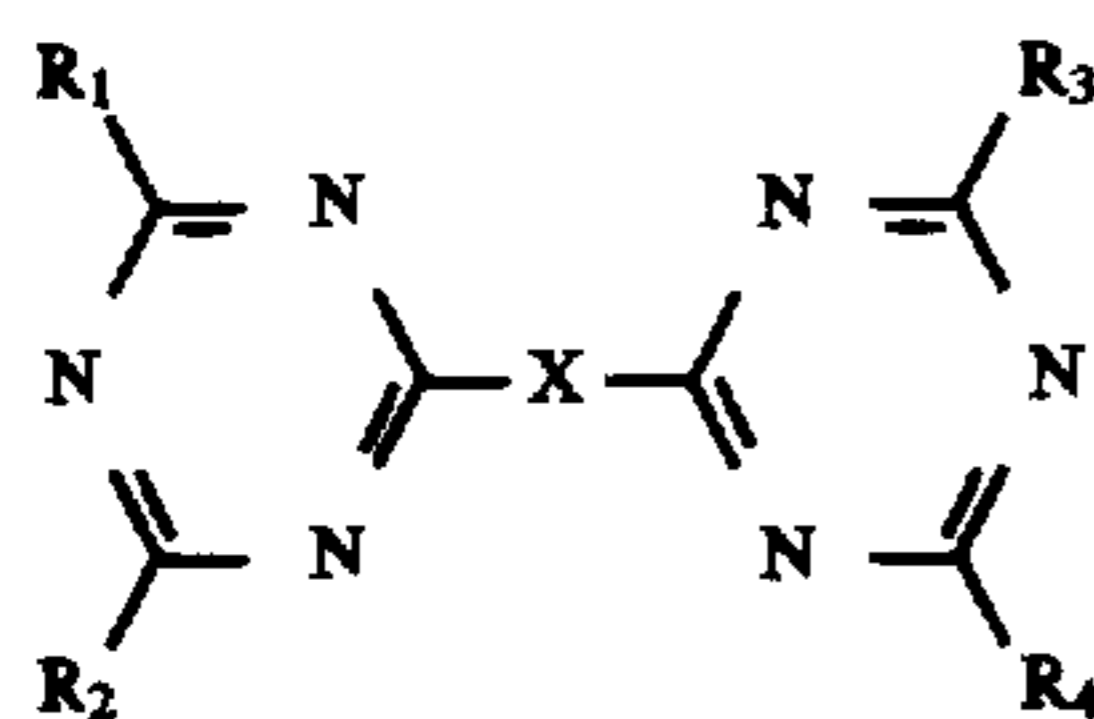
The technical pentaerythritol may also contain some tri and tetra pentaerythritol that is normally formed as by-products during the manufacture of technical pentaerythritol.

The preparation of esters from alcohols and carboxylic acids can be accomplished using conventional methods and techniques known and familiar to those skilled in the art. In general, technical pentaerythritol is heated with the desired carboxylic acid mixture optionally in the presence of a catalyst. Generally, a slight excess of acid is employed to force the reaction to completion. Water is removed during the reaction and any excess acid is then stripped from the reaction mixture. The esters of technical pentaerythritol may be used without further purification or may be further purified using conventional techniques such as distillation.

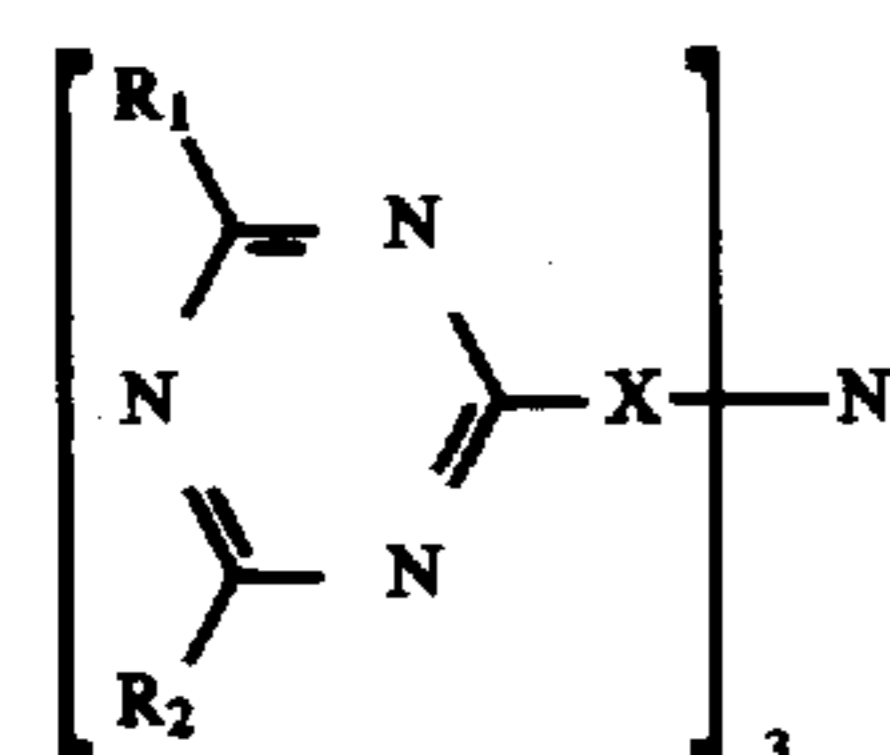
For the purposes of this specification and the following claims, the term "technical pentaerythritol ester" is understood as meaning the polyol ester base oil prepared from technical pentaerythritol and a mixture of C<sub>4</sub>-C<sub>12</sub> carboxylic acids.

As previously stated, to the polyol ester base stock is added a minor portion of an additive mixture comprising a non-sulfur containing triazine derivative and mercapto alkylalcohol.

The non-sulfur containing triazine derivatives are preferably those of the form:

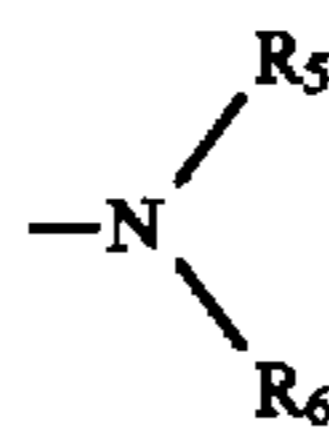


Or alternatively, compound III may also be of the form:



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where R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, R<sub>4</sub> are the same or different and are

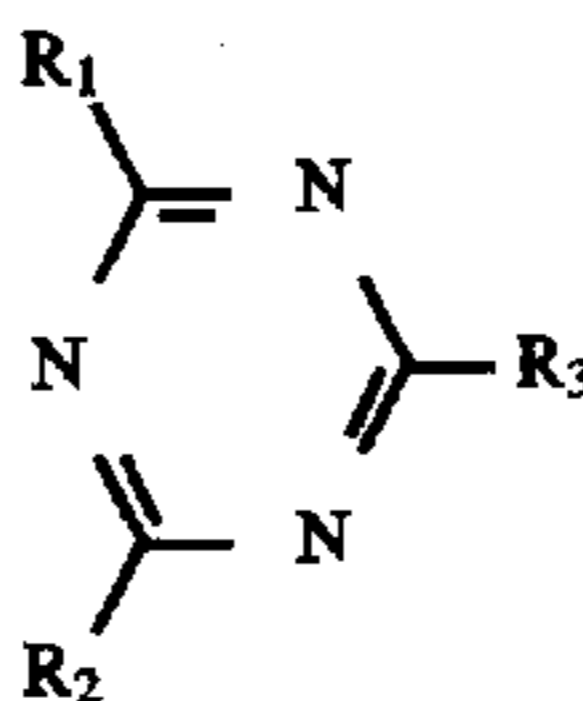


wherein R<sub>5</sub> and R<sub>6</sub> are the same or different and are selected from the group-consisting of C<sub>2</sub> to C<sub>16</sub> branched or straight chain alkyl, aryl-R<sub>7</sub> where R<sub>7</sub> is branched or straight chain C<sub>2</sub> to C<sub>16</sub> alkyl, cyclohexyl-R<sub>7</sub> where R<sub>7</sub> is H or branched or straight chain C<sub>2</sub> to C<sub>16</sub> alkyl. Preferably R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, and R<sub>4</sub> are all the same or different dialkyl amino groups where the alkyl chains are C<sub>4</sub> to C<sub>12</sub>.

For compound III, X is a bridging group selected from the group consisting of piperidino, hydroquinone, NH-R<sub>8</sub>-NH where R<sub>8</sub> is C<sub>1</sub> to C<sub>12</sub> branched or straight chain alkyl and mixtures thereof.

For compound IIIa, X is selected from the group consisting of piperidino, hydroquinone, NH-R<sub>8</sub> where R<sub>8</sub> is C<sub>1</sub> to C<sub>12</sub> branched or straight chain alkyl.

The triazine derivative may also be of the form:



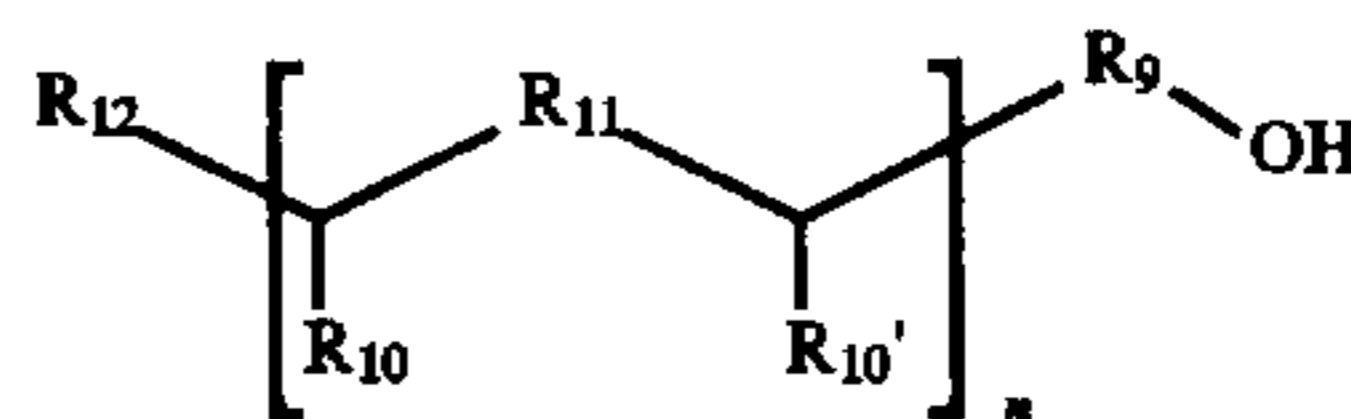
IV

where R<sub>1</sub>, R<sub>2</sub>, and R<sub>3</sub> are identical to the description above. The preferred non-sulfur containing triazines are those of the formula III and IIIa. Those of formula IV are less preferred due to their lower molecular weight which leads to higher volatility and poorer suitability for high-temperature synthetic oil use.

The non-sulfur containing triazine antioxidant is used in an amount in the range 0.1 to 1.2 percent by weight (based on polyol ester base stock), preferably 0.2 to 0.9 percent, most preferably 0.4 to 0.7 percent.

As previously stated, to the synthetic oil base stock is added a minor portion of an additive comprising a mixture of a triazine derivative and a mercapto alkylalcohol.

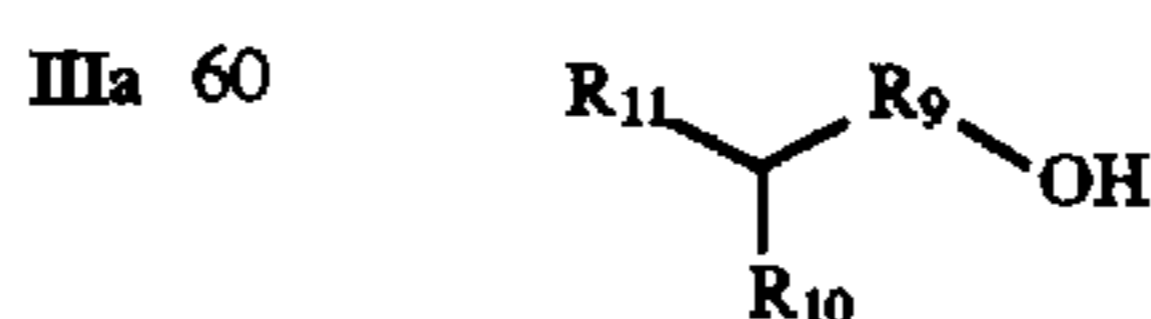
Mercapto alkylalcohols are described by the structural formula:



V

where R<sub>9</sub> and R<sub>10</sub> are C<sub>1</sub> to C<sub>12</sub> alkyl and R<sub>10</sub> and R<sub>10'</sub> are the same or different and are SH or OH and R<sub>12</sub> is C<sub>1</sub> to C<sub>12</sub> alkyl or SH or OH and n is 1 to 4. Additionally, at least 1 of R<sub>10</sub>, R<sub>10'</sub> or R<sub>12</sub> must be SH.

Another form for mercapto alkyl alcohols are described by the structural formula:

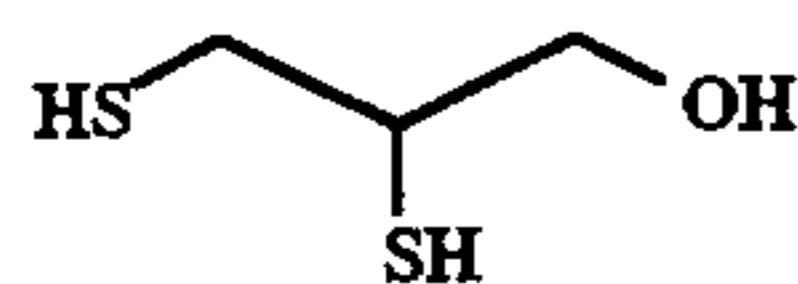


Va

Where R<sub>9</sub> is C<sub>1</sub> to C<sub>12</sub> alkyl, R<sub>10</sub> and R<sub>11</sub> are SH or OH with at least one of R<sub>10</sub> or R<sub>11</sub> being SH.

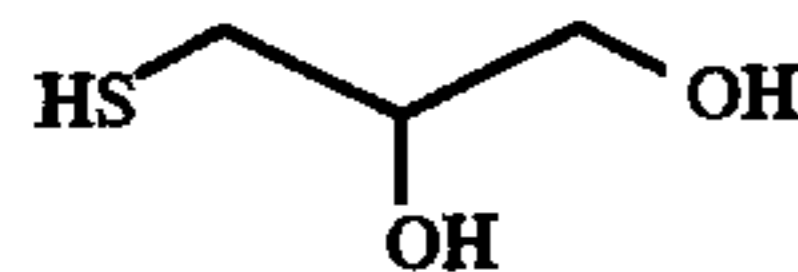
Two examples of preferred MAA compounds are 2,3 dimercapto propanol (VI):

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VI

and 3-mercapto-1,2-propanediol (VII):



VII

The non-sulfur containing triazine antioxidant is used in an amount in the range 0.1 to 1.2 percent by weight, preferably 0.2 to 0.9 percent, most preferably 0.4 to 0.7 percent, while the MAA is used in an amount in the range 50 to 2000 ppm, preferably 100 to 600 ppm, most preferably 200 to 500 ppm.

The non-sulfur containing triazine antioxidant and the mercapto alkyl alcohol are used in a ratio in the range of 2:1 to 100:1, preferably 4:1 to 40:1, most preferably 5:1 to 20:1.

The reduced-deposit oil, preferably synthetic polyol ester-based reduced-deposit oil may also contain one or more of the following classes of additives: antifoamants, antiwear agents, corrosion inhibitors, hydrolytic stabilizers, metal deactivator, detergents and additional antioxidants. Total amount of such other additives can be in the range 0.5 to 15 wt %, preferably 2 to 10 wt %, most preferably 3 to 8 wt %.

Antioxidants which can be used include aryl amines, e.g. phenyl naphthyl amines and dialkyl diphenyl amines and mixtures thereof, hindered phenols, phenothiazines, and their derivatives.

The antioxidants are typically used in an amount in the range 1 to 5%.

Antiwear additives include hydrocarbyl phosphate esters, particularly trihydrocarbyl phosphate esters in which the hydrocarbyl radical is an aryl or alkaryl radical or mixture thereof. Particular antiwear additives include tricresyl phosphate, t-butyl phenyl phosphates, trixylenyl phosphate, and mixtures thereof.

The antiwear additives are typically used in an amount in the range 0.5 to 4 wt %, preferably 1 to 3 wt %.

Corrosion inhibitors include but are not limited to various triazols e.g. tolyl triazole, 1,2,4 benzene triazol, 1,2,3 benzene triazol, carboxy benzotriazole, alkylated benzotriazole and organic diacids, e.g., sebacic acid.

The corrosion inhibitors can be used in an amount in the range 0.02 to 0.5 wt %, preferably 0.05% to 0.25 wt %.

As previously indicated, other additives can also be employed including hydrolytic stabilizers, pour point depressants, anti-foaming agents, viscosity and viscosity index improvers, etc.

Lubricating oil additives are described generally in "Lubricants and Related Products" by Dieter Klamann, Verlag Chemie, Deerfield, Fla., 1984, and also in "Lubricant Additives" by C. V. Smalheer and R. Kennedy Smith, 1967, pp. 1-11, the disclosures of which are incorporated herein by reference.

The additive combinations are useful in ester fluids including lubricating oils, particularly those ester fluids useful in high temperature avionic (turbine engine oils) applications. The additive combinations of the present invention exhibit excellent deposit inhibiting performance and improved oxidative stability as measured in the Inclined Panel Deposition Test.

The present invention is further described by reference to the following non-limiting examples.

#### EXAMPLE 1

This example illustrates the deposit formation performance for the most preferred embodiment of the invention

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by evaluating fully formulated oils in the Inclined Panel Deposit Test ("IPDT"). The additives tested were blended into a finished turbo oil formulation suitable for applications covered by the MIL-L-23699 specifications by using a constant package of additives and basestock. The basestock was a technical pentaerythritol ester made with an acid mixture of C<sub>5</sub> to C<sub>10</sub> commercially available acids. The additive package contained diaryl amine antioxidants, a commonly used metal passivator containing triaryl phosphates, a corrosion inhibitor consisting of alkylated benzotriazole, and a hydrolytic stabilizer. The total concentration of these other additives was 4.342 gms/100 gms polyol ester base stock.

The IPDT is a bench test consisting of a stainless steel panel electrically heated by means of two heaters inserted into holes in the panel body. The test temperature is held at 299° C. The panel temperature is monitored using a recording thermocouple. The panel is inclined at a 4° angle and oil is dropped onto the heated panel near the top, allowing the oil to flow the length of the panel surface, drip from the end of the heated surface and be recycled to the oil reservoir. The oil forms a thin moving film which is in contact with air flowing through the test chamber. Test duration is 24 hours. Deposits formed on the panel are rated on a scale identical to that used for deposits formed in the bearing rig test (FED. Test Method STD. No. 791C, Method 3410.1). Varnish deposits rate from 0 (clean metal) to 5 (heavy varnish). Sludge deposits rate from 6 (light) to 8 (heavy). Carbon deposits rate from 9 (light carbon) to 11 (heavy/thick carbon). Higher ratings (12 to 20) are given to carbon deposits that crinkle or flake away from the metal surface during the test. The total weight of the deposit formed in 24 hours is also measured. In addition, the final viscosity, measured at 40° C., and Total Acid Number ("TAN"), expressed as mg KOH/100 ml, of the used oil are measured after the test is complete, and used as an evaluation of the oxidation of the oil.

Table 1 illustrates the deposition synergistic effect between a series of MAA compounds and triazine compound III, "Triazine", where R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub> and R<sub>4</sub> are all dibutylamino and X is piperidino. The MAA compounds used were:

Compound VI: 2,3-dimercapto propanol

Compound VII: 3-mercapto-1,2-propanediol

The concentration of the triazine is 0.6 gms/100 gms basestock in all cases.

TABLE 1

MAA Compound	Triazine	MAA Concentration	Deposit Rating	Deposit Weight
None	None	N/A	4.3	0.24 gms
None	0.6%	None	3.9	0.25 gms
VI	None	0.05%	2.0	0.07 gms
VI	0.6%	0.05%	1.6	0.00 gms
VII	None	0.05%	4.7	0.44 gms
VII	0.6%	0.05%	3.5	0.18 gms

Table 1 shows that the addition of the triazine has little effect on the deposition performance. The addition of compound VI without the triazine present does substantially reduce the deposit rating and weight. However, the addition of the triazine to compound VI, results in a further dramatic reduction: the deposit rating is reduced an additional 20% and the deposit weight is reduced to 0 (100% reduction) within the 0.005 gm accuracy of the measurement. The addition of compound VII actually increases both the

deposit rating and weight from the base cases presented in the first two lines of Table 1. However, the addition of the triazine to compound VII reduces the deposit rating and weight significantly below the use of the triazine alone: an 11% reduction in the rating and a 28% reduction in deposit weight.

### EXAMPLE 2

Measurement of the oxidative degradation of the oil tested in Example 1 were made by measuring the change in viscosity and acid number, TAN, versus the fresh oil.

Table 2 illustrates the oxidative synergisms for the same compounds in the same test by measuring the percent increase in viscosity and the increase in TAN. The decrease in deposit weight, illustrated in Table 1, might be expected to result in increased Viscosity increase or TAN increase. This is due to solubilization of incipient deposits by the oil resulting in a larger concentration of high molecular weight, partially oxidized molecules. However, Table 2 clearly illustrates that no such effect is observed. Viscosity and TAN changes are dramatically lower for these combinations indicating that not only are deposits reduced as shown in Example 1, but incipient deposits and other partially oxidized species are not formed in the same quantities when both the triazine and MAA compounds are present.

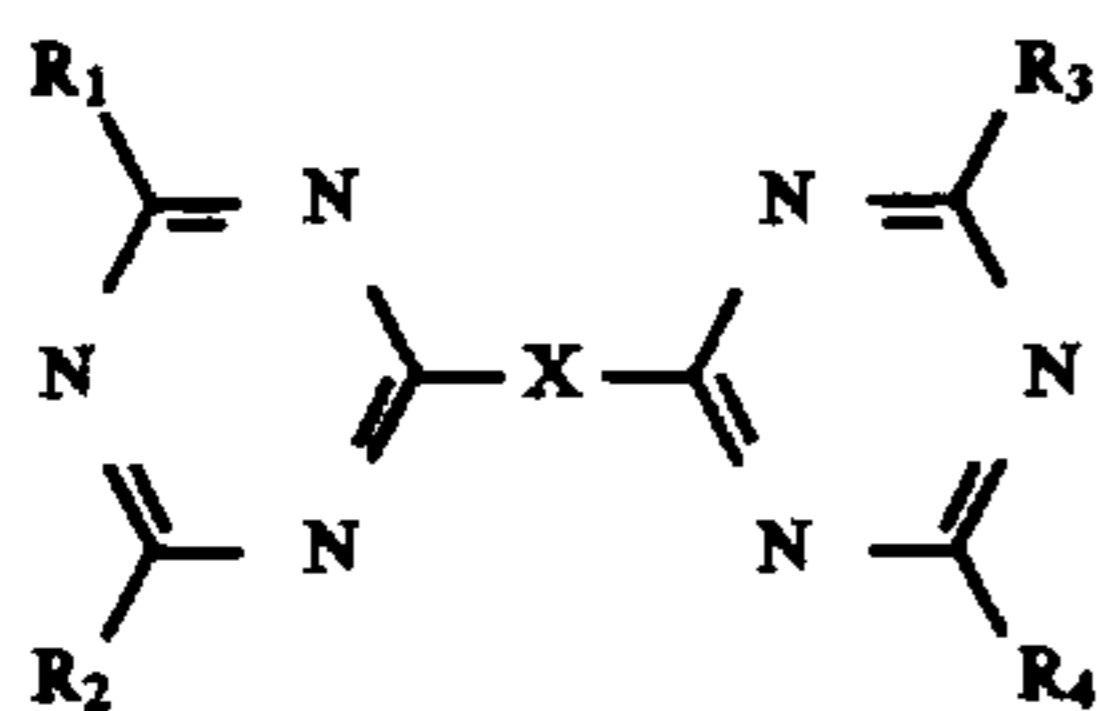
TABLE 2

MAA Compound	Triazine	MAA Concentration	Viscosity Increase	TAN Increase, mg KOH/L
None	None	N/A	101%	14.2
None	0.6%	None	94%	10.5
VI	None	0.05%	4.6%	1.8
VI	0.6%	0.05%	<1%	0.5
VII	None	0.05%	124%	14.3
VII	0.6%	0.05%	34.2%	3.7

Significant improvements in Viscosity and/or TAN increase are observed for combinations of compounds VI and VII with triazine over any formulation without both compounds present. For compound VI, the combination results in no increase in viscosity versus 4.6% for compound VI without the triazine, and a reduction in TAN increase of 72% for the combination over the use of compound VI alone and 95% over the use of the triazine alone. For compound VII, the combination reduces the Viscosity increase by 73% over the use of compound VII alone and 64% over the use of the triazine alone; the TAN increase is reduced by 74% for the combination versus compound VII alone and 65% for the combination versus the use of the triazine alone.

What is claimed is:

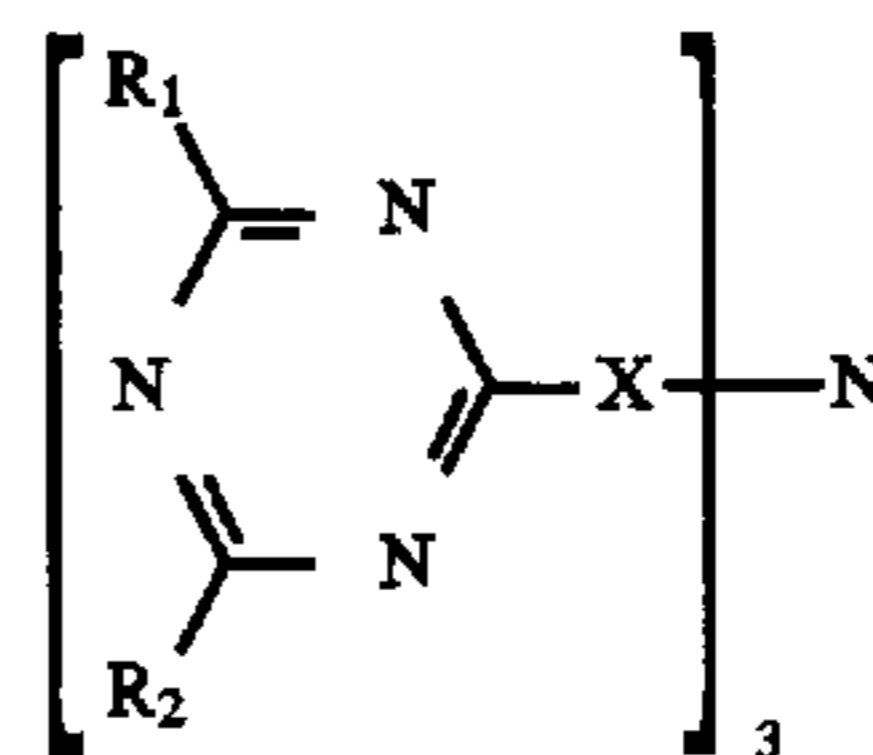
1. A turbo oil composition exhibiting enhanced resistance to deposition and improved oxidative stability, said turbo oil lubricant composition comprising a synthetic ester based base stock selected from diesters and polyol esters and 0.1 to 1.2 percent by weight of a non-sulfur containing substituted triazine derivative of the formula:



III

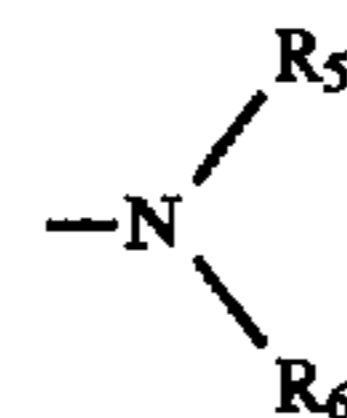
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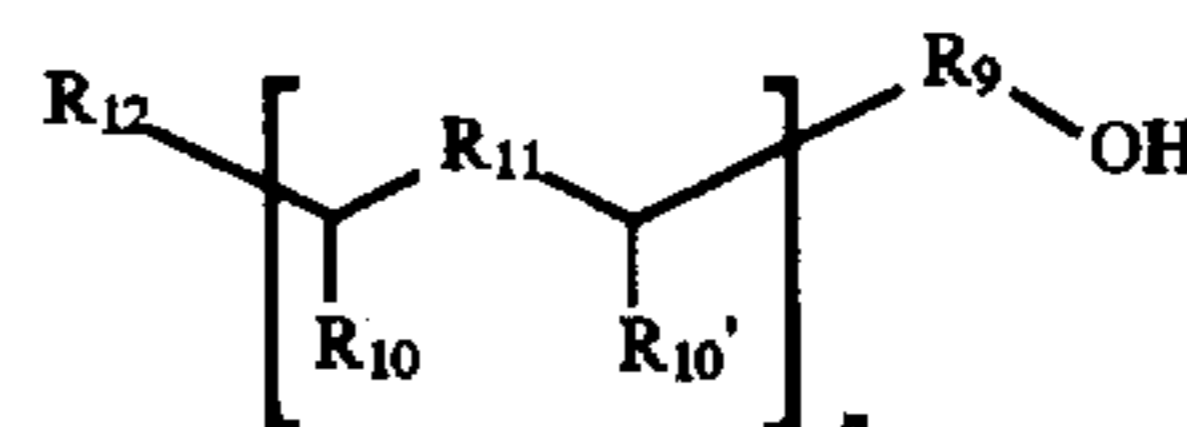


IIIa

where  $R_1, R_2, R_3, R_4$  are the same or different and are

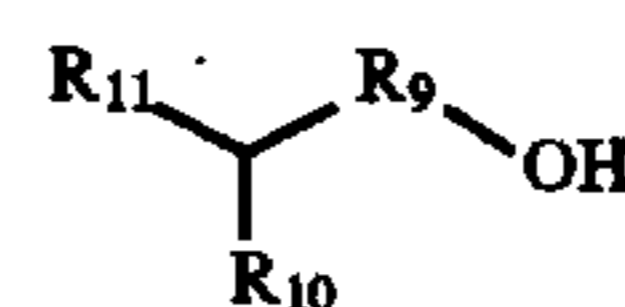


wherein  $R_5$  and  $R_6$  are the same or different and are selected from the group consisting of  $C_2$  to  $C_{16}$  branched or straight chain alkyl, aryl- $R_7$  where  $R_7$  is branched or straight chain  $C_2$  to  $C_{16}$  alkyl and cyclohexyl- $R_7$  where  $R_7$  is H or branched or straight chain  $C_2$  to  $C_{16}$  alkyl; and wherein in formula III X is a bridging group selected from the group consisting of piperidino, hydroquinone, and  $NH-R_8-NH$  where  $R_8$  is  $C_1$  to  $C_{12}$  branched or straight chain alkyl, and in formula IIIa X is selected from the group consisting of piperidino, hydroquinone and  $NH-R_8$  where  $R_8$  is  $C_1$  to  $C_{12}$  branched or straight chain alkyl; and 50 to 2000 ppm of a mercapto alkyl alcohol as represented by the structural formula:



V

where  $R_9$  and  $R_{11}$  are  $C_1$  to  $C_{12}$  alkyl and  $R_{10}$  and  $R_{10}'$  are the same or different and are SH or OH and  $R_{12}$  is  $C_1$  to  $C_{12}$  alkyl or SH or OH,  $n$  is 1 to 4, and wherein at least 1 of  $R_{10}, R_{10}'$  or  $R_{12}$  must be SH, or



Va

wherein  $R_9$  is  $C_1$  to  $C_{12}$  alkyl,  $R_{10}$  and  $R_{11}$  are SH or OH with at least one of  $R_{10}$  or  $R_{11}$  being SH.

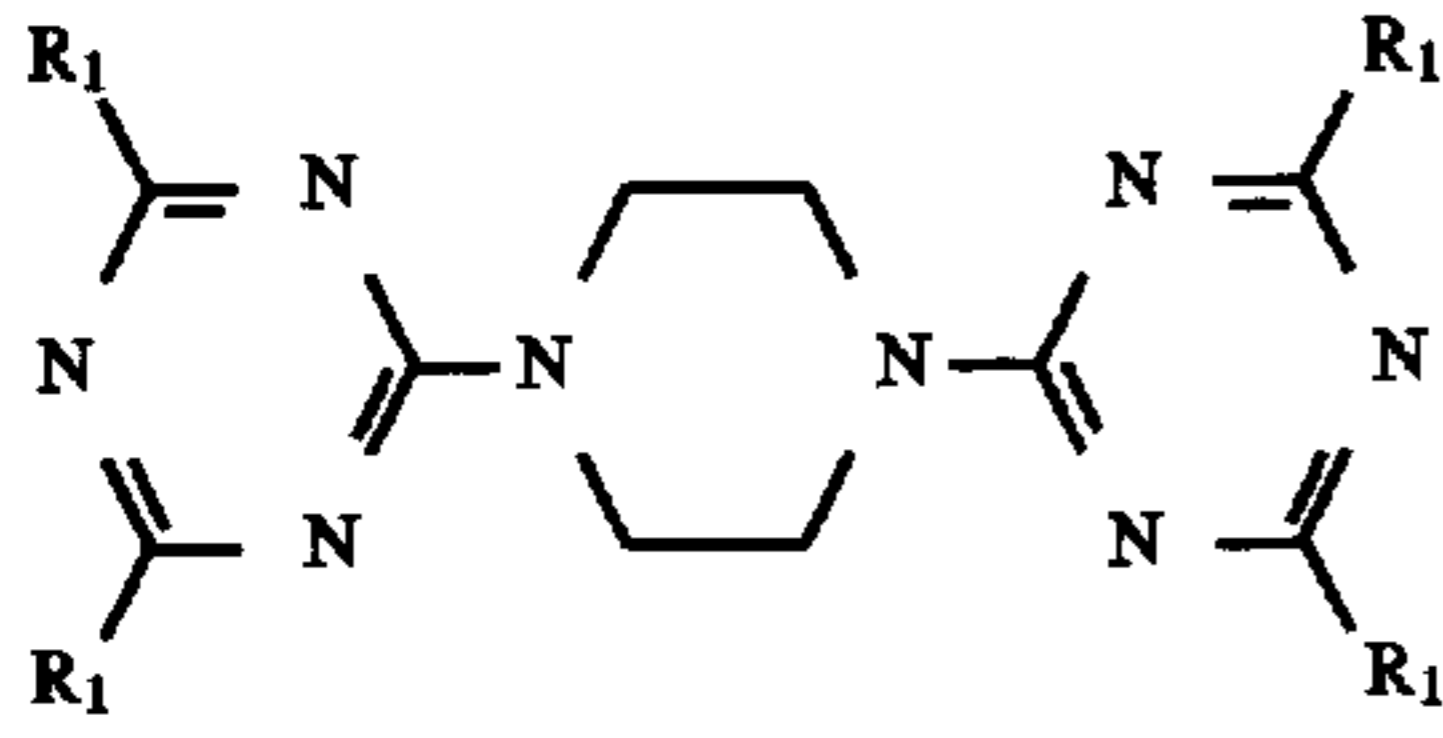
2. The turbo oil lubricant composition of claim 1 wherein the synthetic polyol ester based base stock is the esterification product of an aliphatic polyol containing 4 to 15 carbon atoms and from 2 to 8 esterifiable hydroxyl groups reacted with a carboxylic acid containing from 4 to 12 carbon atoms.

3. The turbo oil lubricant composition of claim 2 wherein the synthetic polyol ester based base stock is the esterification product of technical pentaerythritol and a mixture of  $C_4$  to  $C_{12}$  carboxylic acids.

4. The turbo oil lubricant composition of claim 1 wherein the non-sulfur containing substituted triazine derivative and mercapto alkyl alcohol are used in a ratio in the range of 2:1 to 100:1 by weight.

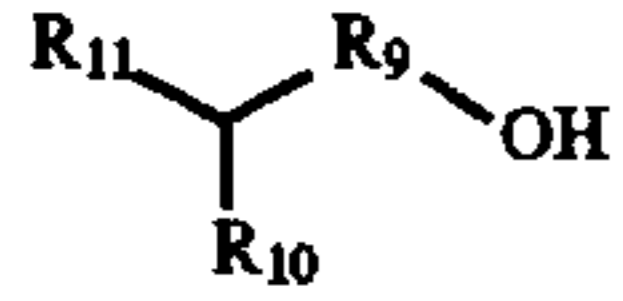
5. The turbo oil lubricant composition of claim 1 where the substituted triazine is of the formula:

9



where  $R_1$  is dibutylamino.

6. The turbo oil lubricant composition of claims 1, 2, 3, 4 or 5 wherein the mercapto alkyl alcohol is represented by the structural formula:



Va 15

10

wherein  $R_9$  is  $C_1$  to  $C_{12}$  alkyl,  $R_{10}$  and  $R_{11}$  are SH or OH with at least one of  $R_{10}$  or  $R_{11}$  being SH.

7. The turbo oil lubricant composition of claim 6 wherein  $R_{10}$  and  $R_{11}$  are SH and  $R_9$  is  $C_1$  to  $C_4$ .

8. The turbo oil lubricant composition of claim 7 wherein  $R_9$  is  $C_1$ .

9. The turbo oil lubricant composition of claim 6 wherein  $R_{11}$  is SH,  $R_{10}$  is OH and  $R_9$  is  $C_1$  to  $C_4$ .

10. The turbo oil lubricant composition of claim 9 wherein  $R_9$  is  $C_1$ .

\* \* \* \* \*